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WORKSHOP ON DOSIMETRY FOR RADON AND RADON DAUGHTERS,

OAK RIDGE NATIONAL LABORATORY, APRIL 12-13, 1977

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CONTENTS

WORKSHOP ON DOSIMETRY FOR RADON AND RADON DAUGHTERS

Edited by
J. E. Turner, C. F. Holoway, and A. S. Loebl

ABSTRACT

The two-day subject Workshop was held at Oak Ridge National Laboratory in April 1977. Approximately 20 persons from several laboratories and agencies participated. Emphasis was placed on the dosimetry for radon and daughters, rather than on monitoring and instrumentation. The objectives of the meeting were to exchange scientific information, to identify problem areas in radon-daughter dosimetry, and to make any observations or recommendations by the participants through issuance of this report. The discussion topics included the history of dosimetry for radon and daughters, human data, aerosols, deposition and movement in the respiratory tract, dose calculations, dose-to-working-level-month (WLM) conversion factors, animal experiments, and the development of regulations and remedial criteria for reducing population exposures to radon daughters. This report contains a summary of Workshop discussions plus individual statements contributed by several of the participants. The outstanding problem areas from the standpoint of dosimetry appear to involve the appropriate lung organ mass to be used (average lung-tissue dose vs. high-level local dose); recognition of the discrete, rather than continuous, structure of the mucus; lack of knowledge about lung clearance; the variability of dose with the degree of disequilibrium and the unattached fraction of radon daughters for a given WLM; and questions about the character of the mine atmospheres actually breathed in the older mines from which much of the epidemiological information originates. The development of criteria for taking remedial action to reduce exposures involves additional concerns of basing long-term risk assessment on short-term sampling and applying WLM data for miners to general populations.

I. INTRODUCTION

J. E. Turner, C. F. Holoway, and A. S. Loebl

1. Workshop Objectives

A Workshop on Dosimetry for Radon and Radon Daughters was held by the Health Physics Division (now Health and Safety Research Division) at Oak Ridge National Laboratory, April 12-13, 1977. A number of experts in various aspects of radon-daughter dosimetry were invited to participate. Several attendees agreed in advance of the meeting to act as discussion leaders on specific subjects, which were covered in about the same order as they appear in Sections II and III of this report. The Workshop was conducted informally.

Emphasis at the Workshop was placed on dosimetry and, particularly, on problem areas concerned with converting measurements of an inhaled atmosphere into estimates of dose and risk. Instrumentation and personnel and area monitoring were not covered. The discussions were guided to try to identify where the greatest uncertainties arise in converting information about exposure into estimates of dose. What things are generally agreed on? What things are not? What implications do answers to these questions have for the validity of standards and regulations? What research is needed? Because the Workshop concentrated on technical problems of dosimetry, it supplemented the meeting at the Health and Safety Laborato[illegible] in February 1977 (Breslin 1977),* with emphasis on monitoring, and th[illegible] [illegible]r Energy Agency's conference at Elliot Lake, Canada, in Octobe[illegible] (OECD 1977), with emphasis on mine problems. Oak Ridge National Laboratory is participating in the radiological surveying of active and inactive uranium mill-tailings sites and in the development of criteria for decommissioning land contaminated with radium. These activities, in part, provided a stimulus for organizing the Workshop and developing its technical themes.

The Workshop fulfilled two principal objectives. First, it provided an opportunity for the exchange of information and points of view on both technical and philosophical questions about radon-daughter dosimetry. Second, the Workshop resulted in the present report, which in a sense represents a state-of-the-art assessment of problems in dosimetry for radon and radon daughters. Following W. S. Snyder's review in Section II, the editors have tried to summarize the two days' discussions in Section III. Several of the participants supplied individual statements, which are collected in Section IV. The Workshop findings and recommendations are given in Section V.

This report has been circulated in draft among the Workshop participants and several other experts not in attendance. The editors are

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*References are given at the end of each section.

grateful for the help they received both at the Workshop and in the preparation of this report. While comments and suggestions from a number of persons have been freely incorporated into this report, the editors take responsibility for its shortcomings.

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Breslin, A. J., 1977, ed., Radon Workshop - February 1977, Report HASL-325, National Technical Information Service, U. S. Department of Commerce, 5285 Port Royal Road, Springfield, VA 22161.

OECD, 1977, Personal Dosimetry and Area Monitoring Suitable for Radon and Daughter Products, Proc. NEA Specialist Meeting, Elliot Lake, Canada, 4-8 October 1976, Nuclear Energy Agency, Organization for Economic Cooperation and Development, 2, rue André-Pascal, 75775 Paris Cedex 16, France.

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3. Some Physical Data on Radon and Radon Daughters (See also Section IV-3.)

The principal pathway by which $^{222}_{86}Rn$ decays into stable $^{206}_{82}Pb$ is shown in Fig. I-1. The corresponding decay energy and half-life of each nuclide, as well as its atomic number Z and atomic mass number A, are shown. Alpha decay is represented by the arrows pointing downward toward the right and beta decay by the vertical arrows.

The principal hazard from radon daughters is the deposition in the respiratory tract of the short-lived alpha emitters, RaA and RaC'. The ranges of the particles emitted by these nuclei are about 45 μ and 70 μ in lung tissue (Harley and Pasternack 1972).

The working level (WL) was introduced by Holaday et al. (1957) to give a simple physical measure of the hazard from radon-daughter exposure. One working level is defined as any combination of short-lived daughters (RaA, RaB, RaC, RaC') in one liter of air that will result in the emission (by them) of 1.3×10^5 MeV of alpha-particle energy. This is the amount of alpha energy associated with the short-lived daughters in secular equilibrium with 100 pCi of $^{222}_{86}Rn$.

To see this equivalence, we note that 100 pCi corresponds to a decay rate of 3.7 disintegrations per second. Since the decay rate for a radionuclide is equal to $(0.693/\tau)N$, where τ is its half-life and N is the number of atoms present, we have, for 100 pCi,

$$N = \frac{3.7\ \tau}{0.693}$$

with τ expressed in seconds. Table I-1 shows the number of atoms of the short-lived daughters in secular equilibrium with 100 pCi of $^{222}_{86}Rn$. The decay of RaC' is treated as though it were instantaneous in specifying the activity. Each RaA atom will result in the emission of its own alpha particle plus the one from RaC'. The total alpha energy is thus seen to be 1.3×10^5 MeV.

ORNL-DWG 78-6142

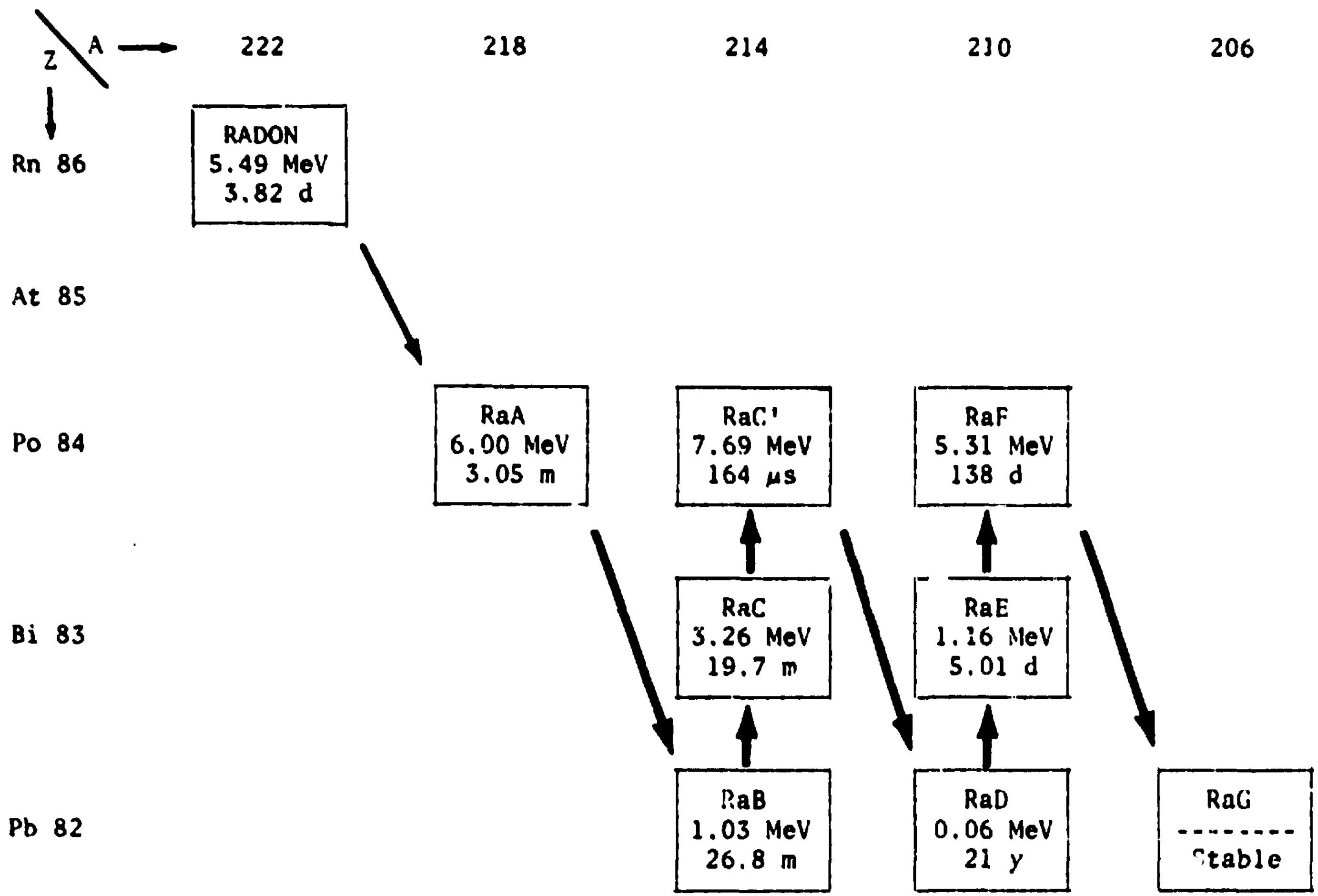

Fig. I-1. Radon and radon daughters.

Table 1-1. Potential alpha-particle energy from short-lived daughters in secular equilibrium with 100 pCi of $^{222}_{86}Rn$

Nuclide	Half-life, τ (sec)	Number of atoms, N	Alpha energy E_α (MeV)	NE_α (MeV)
RaA	183	977	6.0 + 7.7	1.34×10^4
RaB	1608	8585	7.7	6.61×10^4
RaC-RaC'	1182	6311	7.7	4.86×10^4
				$12.81 \times 10^4 =$ 1.3×10^5 MeV

The working level specifies concentration; the working level month (WLM) is used to specify exposure. Inhalation of radon daughters at a concentration of 1 WL for one month (168 hours of working time) results in an exposure of 1 WLM. The cumulative working level month (CWLM) is also used to give accumulated exposure. Breathing rate is not specified in the definition of the WLM.

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II. HISTORICAL SUMMARY OF DOSIMETRY FOR RADON AND RADON DAUGHTERS--CONCEPTS AND PROBLEMS

W. S. Snyder

First of all, it should be emphasized that a "history" of what was essentially an action by committees can only be a personal history. Different members of the committee may agree for quite different reasons which are not apparent from the minutes or even from the discussion. Thus, this account will be inevitably a rather personal account.

During the years preceding the issuance of the reports by the International Commission on Radiolcgical Protection (ICRP) and the National Council on Radiation Protection and Measurements (NCRP) Committees 2 (1959), my colleagues and I were gathering materials on many radionuclides, and among them was ^{222}Rn. One had the value proposed by Evans and Goodman (1940) for ^{222}Rn plus daughters and a value computed by K. Z. Morgan for ^{222}Rn without daughters (1954); and these two values, both based on a dose to lungs, differed by several orders of magnitude. We were aware of the many factors in the mines that might influence the dose at least in a general way, say, attachment of daughters, ventilation and presence of other radionuclides, and the contribution of dose from external sources. We were also aware that the problem was of some importance, for already Duncan Holaday was voicing concern over the incidence of lung cancer. A review of the previously recommended values is given by C. G. Stewart and S. D. Simpson (1964); and this includes a discussion of various uncertainties present in the minds of several committee members, i.e., were alpha particles to have a quality factor of 20 or of 10, was the recommendation based on a 40-hour week or was it for continuous exposure, etc.

I do not want to give the impression that I understood each of these subjects as much as we understand them today. In particular, the statistical analysis of the deaths of uranium miners from lung cancer was still several years in the future. Rather, we were grasping for whatever seemed to offer some clue to the dosimetry of the mining situation.

One of the papers we studied was that by Shapiro (1956 and 1954). This represented a portion of his doctoral dissertation at the University of Rochester; and although many assumptions in use at that time were changed later, I quote a paragraph from his section of Comments:

> "Average radiation dosage to the epithelial lining of the larger bronchi is more difficult to estimate from experimental data. Extrapolation of the results of the dose experiments to human subjects suggests that this dosage may reach 10 times the dosage to the lung as a whole.
>
> "Since even in air containing a considerable amount of large particle dusts the radon daughters seem to be largely deposited on small particles, the deposition of radon daughters still appears to be largely by diffusion. Therefore, the average dose to the bronchial tubes probably does not depend very strongly on the breathing pattern of the individual being exposed as long as the air flow through the lung passageways is laminar. In one experiment the activity deposited in plastic tubes approximately 2 mm in diameter was measured as a function of minute volume. An average decrease of minute volume of 59% decreased the activity deposited in the tube by only 29%. Accordingly, the tracheal dosages reported in this paper may give a fair indication of the dosages imparted to tubes of similar size in the human respiratory tract. They suggest that average radiation dose to the human bronchi may be a few times higher than to the lungs as a whole and, therefore, may reach or exceed 15 mrep per week."

This is one of the few experimental papers which discusses and offers suggestions about the dosimetry of the human instead of stopping with the "results observed in the dog." In any case, this experiment indicated what might be the tissue receiving the highest dose, and it was not the lung as a whole.

In those days perhaps we were more impressed by the tissue receiving the highest dose than we might be today. For example, we tried to estimate the maximum dose in the sections of the gastrointestinal tract, and any radiation penetrating the skin was generally listed as "whole body dose." We did not try to estimate the dose from a "hot particle" or for a single track of an alpha particle; but, still, if a gram mass or more of tissue were irradiated at a higher level than the average, we tended to consider this dose rather than the average dose in deriving a maximum permissible concentration. Partly, this stems from the fact that neither the NCRP nor the ICRP has ever seen fit to define what it means by an organ or to declare that only doses to a selected list of organs are to be considered. Likewise, they fail to emphasize that it is the average dose to the organ that is of concern except for very special radiation exposure situations. In any case, we had rather strong experimental support for the position that the maximum dose probably occurred in the epithelial lining of the larger bronchi.

Another paper which influenced our thinking was by Chamberlain and Dyson (1956); for this study, in a sense, supplied the quantitative basis for the qualitative results already mentioned. Chamberlain and Dyson constructed a model of the human trachea and included about 4 cm of the upper bronchi. Through this they drew a steady stream of air and so studied the deposition of activity on the wall of the model under controlled conditions. They gave graphs showing this measured activity as a function of the breathing rate and of the degree of attachment of the radon daughter atoms to condensation nuclei. From this data, they calculated the dose rate to the tissue.

It is easy to criticize this experiment, and the authors only partially answered some of these objections. For example, they coated the walls of the model trachea with a sticky solution to see if the character of the surface would greatly perturb the deposition. This did not significantly affect the results. Perhaps the greatest objection might be that the steady air flow through the model is not representative of the breathing pattern with its fairly abrupt reversals of direction and variable rates all of which may well produce turbulence. A turbulent condition probably affects the deposition, since they noted that deposition was high near the larynx, and this they attributed to turbulence caused by the connection of the tube.

There is not much more to say. We presented these results to the Committees of ICRP and NCRP and they agreed. Actually there was a dose calculated by K. Z. Morgan for radon without daughters, and a formula was devised which would produce this dose if the degree of attachment approached 100%. The dose for an attachment coefficient of 10% is given as typical of unfiltered room air.

Thus, this dose level was largely a result of experiment. How has it fared in the years since it was adopted? In most of the data from the Colorado Plateau, it gives one more variable which largely would have to be guestimated since few measurements were available. The analysis of the data took a quite different turn and has been expressed nearly always in terms of μCi-months of exposure. Since the exposure data nearly always extend over periods of years when ventilation patterns and other conditions are changing, it may be best not to try to guess the degree of attachment.

The various lung models that have been proposed seem to agree with the main thesis that the higher doses, if not the highest, are probably those in the upper bronchi.

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III SUMMARY OF DISCUSSIONS

J. E. Turner and C. F. Holoway

1. Human Experience

Archer presented a review of epidemiological data for uranium miners. The reader is referred to Section IV-1 of this report for the summary he has contributed. Lung-cancer incidence in excess of normal numbers has been analyzed according to the cumulated exposure of workers and their height and smoking habits. The data show a higher incidence in shorter workers and in workers who smoke. Presumably, tall and short men do about the same amount of work and, therefore, consume the same amount of oxygen. Because of smaller lung volume, a shorter worker on the average inhales and exhales more often, trapping more airborne radon daughters than a tall worker in the same atmosphere. Cigarette smoking is often accompanied by changes in lung clearance, observed metaplasia in the bronchi, and probable promoting effects. Data on the incidence of emphysema and chronic bronchitis among uranium miners (who are exposed to "free silica" as well as to radon daughters) also show similar patterns with respect to height and smoking habits. With the uranium miners, the cancer incidence in the "short, heavy-smoker" category is about 60-65 times that in the "tall, nonsmoker" category.

The slopes of the excess lung cancer incidence curves become steeper by a factor of about 3 or 4 at lower dose rates. While the latent period is found to depend strongly on the age at which work in the mine began, it is not yet known whether it changes with working level month (WLM).

It was pointed out that the new ICRP recommendations, which will consider total risk rather than risk to critical organs, will probably involve an independent evaluation of the U. S. and Czech data based on the WLM.

A recurring theme of the Workshop was struck on the relative merits of using the WLM or the absorbed dose as a measure of risk. While the WLM provides a very useful physical measure of hazard from radon daughters, its critics point out that the dose implied by a given WLM can vary by one or two orders of magnitude. The variation depends critically upon the unattached fractions of the daughters RaA, RaB, and RaC, which, in turn, depend on ventilation rate, particle-size distribution, and other factors. Generally, published rad/WLM values seem to have decreased through the years to around unity, or less, today. The conversion factors seem to fall into two groups--for mines, below 1; and for outdoor air, around 3. The additional observation was made that ventilation rates have changed over the years, and, therefore, the older WLM data may not be representative of today's conditions. The reader is referred to Sections III-3, 4; IV-4, 5; and V for additional comments on lung dosimetry and the WLM.

2. Radon Daughters and Aerosols

The first step in translating measurements of airborne radon and radon daughters into estimates of dose is understanding the physical characteristics of the inhaled radionuclides. The noble gas $^{222}_{86}Rn$ atom can diffuse through material in which its parent $^{226}_{88}Ra$ atom was trapped and escape into the atmosphere. Airborne radon remains unattached and decays by alpha-particle emission, producing $^{218}_{84}Po$ (RaA) with a recoil energy of 101 keV. The recoiling polonium atom is partially stripped of its electrons by the departing alpha particle and by collisions with air molecules, a positively charged polonium ion usually being left. This ion remains free until it decays radioactively, reacts with oxygen, forms the nucleus for a cluster of water molecules, or attaches to aerosol particles (Raabe 1969). Combinations of these processes can occur, one example being the attachment of water-atom clusters to airborne particulates. A similar analysis applies to the daughter RaB, except that its fate can be influenced by the physical state of the RaA parent.

In principle, deposition of radon daughters in the respiratory tract is determined by the aerosol particle-size distribution and the distribution of attached and unattached daughters. These distributions, in turn, depend in a complicated way on such factors as the degree of radioactive disequilibrium among radon and its daughters, humidity, the chemical composition of the aerosol, and the presence of surfaces and boundaries in the area. In general, higher ventilation rates favor a greater degree of radioactive disequilibrium. The influence of some of these factors, together with general characterizations of aerosol number concentrations in indoor and outdoor atmospheres and in mines, has been summarized by Fry (1976).

Raabe (1969) has shown that, over a wide range of values of parameters, the attachment rate of radon daughters to aerosols is proportional to the surface area of the aerosol particles. Concentrations of the highly diffusible radon-daughters in most cases of practical interest are low ($\sim$1 ion/10^3 aerosol particles). Diffusion gradients are not established, and adsorption can be described as a random surface interaction in terms of surface area alone. (This circumstance may not be true for the 56-second half-life thoron.)

Based on this principle, it is possible to estimate the attached fraction of radon daughters and the size distribution of particles to which they attach (Raabe 1969). Conversely, if one knows the attachment, then radon-daughter measurements can be used to determine the aerosol distribution. Knowledge of the size distribution and number concentration of aerosols appears to give a sufficient basis for estimating the unattached fraction of radon daughters. It was noted that more measurements should be made to verify these findings.

For additional information the reader is referred to Raabe's contribution in Section IV-3.

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Fry, R. M., 1976, "Radon and Its Hazards," p. 13 in Personal Dosimetry and Area Monitoring Suitable for Radon and Daughter Products, Proc. NEA Specialist Meeting, Elliot Lake, Canada, 4-8 October 1976, Nuclear Energy Agency, 2 rue Andre-Pascal, 75775 Paris Cedex 16, France.

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3. Dose in the Respiratory Tract

Because pathological studies of tumors have indicated a predominantly bronchogenic origin, radon-daughter dosimetry is concerned with the epithelial lining in the bronchial tree. The factors to be evaluated in estimating dose include the deposition of activity in the respiratory tract, its subsequent movement and clearance, the biological targets at risk, and the calculation of the absorbed energy and its spatial distribution. Each of these factors was discussed individually. Most lung models use diffusion equations to calculate deposition in the bronchial tree, although the flow is not strictly laminar. It was generally agreed that inadequacies in treating deposition can probably be corrected by simple modification of the Gormley-Kennedy equations such as that proposed by Martin and Jacobi (1972).

The calculation of average dose and the physics of alpha-particle penetration are more precisely defined than the physiological and pathological parameters. The numerical value of the absorbed dose, determined as an average energy deposited in a volume from many particles, does not depend strongly on the details assumed about source and target. Various published calculations of absorbed dose do not differ greatly.

However, the question of high-level "local" doses versus "average" lung dose still remains unsettled. Evans (1977) points out that the 20-fold or higher local doses apply to only 0.5 g of tissue--about 0.005 of the total lung mass. As brought out by Snyder in Section II, the ICRP lung model in its present state is not suited for radon (and other noble gases). While the model adequately describes the deposition of particulate matter in the lungs, it does not describe the deposition of ions. In addition, its clearance modes and constants are not detailed enough to give accurately the dose from short-lived activity.

Where are the target cells located and what are the critical sites? All dosimetric models identify the nuclei of the basal cells of the bronchial epithelium as the relevant biological target. However, some uncertainties still exist. Gastineau, Walsh, and Underwood (1972) measured the thicknesses of the epithelium and the depths of the basal-cell layers in the lobar and segmental bronchi from normal human specimens. They found that some regions where tumors are reported are outside the range of the radon-daughter alpha particles. However, they did note areas of metaplasia that were thin enough to permit penetration of an alpha particle into the basal-cell layer, a factor that may account for the greater incidence of lung cancer among miners who smoke. The findings may also indicate that some daughters are soluble and penetrate to the basal-cell layer.

The remaining factor listed at the beginning of this section, lung clearance, probably introduces the greatest uncertainty in calculating dose. Little data exist on mucous production and clearance rates. Mucociliary clearance has been studied by Van As (1977) and Sturgess (1977). Electron-microscope studies indicate that the mucous layer exists in discrete parts, rather than in a continuous blanket over the epithelium as is usually assumed in calculating dose. (However, the preparation of the samples for scanning might contribute to the breakup of the mucous blanket.) The mucous secretions also appear to undergo random motion. Additional uncertainties arise because clearance rates are different in different areas of the respiratory tract. For example, there may be little clearance at bifurcations; removal rates are not well known; clearance rates may be quite different between smokers and nonsmokers. Because of the short half-life of radon daughters, a maximum dose of twice that estimated for uniform deposition can exist if clearance is stopped. However, this factor does not take into account any additional effect of accumulation or very high doses in small localized areas.

In summary, we appear to have a fairly satisfactory aerodynamic lung model for dose calculations but a poor physiologic one for radon daughters.

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Martin, D., and Jacobi, W., 1972, "Diffusion Deposition of Small-Sized Particles in the Bronchial Tree," Health Phys. 23, 23.

Sturgess, J. M., 1977, "The Mucous Lining of Major Bronchi in the Rabbit Lung," Am. Rev. Resp. Dis. 115, 819.

Van As, A., 1977, "Pulmonary Airway Clearance Mechanisms: A Reappraisal," Am. Rev. Resp. Dis. 115, 721.

4. Dose and Working-Level-Month Conversion Factors

Table III-1 is taken from a recent summary by Fry (1976) of radon dose-conversion factors published by several investigators. Specifically, the numbers give the absorbed dose to the nuclei of the basal cells in the segmental bronchi. Various numerical factors assumed in the calculations are also shown in the table. The dosimetry has also been reviewed by Nelson and Parker (1974). They propose that a "much-rounded value of 10 rad/WLM could be used as a guide to cancer related dose, probably to within a factor of 3, but possibly to not better than a factor of 5 or so, for most of the expected conditions of exposure to mine air." The BEIR Report (1972) considers 1 rad/WLM as the probable upper limit for a uniform dose, with perhaps 0.5 rad/WLM for miners. Jacobi (1976) has suggested 0.3 to 0.5 rad/WLM for inhalation by miners. All dose calculations and recommendations for occupational exposure to radon and radon daughters published through 1972 have been summarized by Cross, Bloomster, Hendrickson, and Nelson (1974).

The last column in Table III-1 shows the dose conversion factors. With such a wide range of possible values for the variables, the dose conversion factors vary by more than an order of magnitude. The high value of Haque and Collinson is due to the large values used for unattached RaA, RaB, and RaC (35%, 5.8%, 7.7%). Table IV-1 in Section IV-3 indicates that such high unattached fractions are not likely to occur in practice.

Variations depend to a large extent on the unattached fraction, the degree of disequilibrium, aerosol characteristics, and details of the particular lung model used, particularly the depth assumed for the basal-cell nuclei. Aside from differences that arise because calculations apply to different situations, however, a basic shortcoming of the working-level-month concept is evident. By definition, the WLM depends only on the potential alpha-particle energy and is independent of its

Table III-1. Dose conversion factors for radon and daughters

Reference	Daughter distribution Rn: RaA: RaB: RaC	Disequilibrium factor	Atmosphere	Remarks	rad/WLM
Altshuler, B., Nelson, N., and Kuschner, M., 1964, Health Phys. 10, 1137.	10: 9: 6: 4 9% free RaA	0.58	Mine	Breathing: Nose Mouth	 1.4 3.5
Jacobi, W., 1964, Health Phys. 10, 1163.	10: 10: 10: 10 25% free RaA	1.0	Outdoor		2.7
	10: 10: 6: 4 25% free RaA	0.57	Indoor		3.0
Haque, A. K. M. M., and Collinson, A. J. L., 1967, Health Phys. 13, 431.	10: 9: 5: 3.5 35% free RaA	0.49	Indoor	Cell Nuclei Depth: 60μm 30μm	 1.4 9.9
Jacobi, W., 1972, Health Phys. 23, 3.	Average conditions 1-2% free RaA		Clean air		0.38-1.0
Harley, N. H., and Pasternack, B. S., 1972, Health Phys. 23, 771.	10: 10: 10: 10 4% free RaA	1.0	Mine		0.24
	10: 6: 3: 2 4% free RaA	0.29			0.36
	10: 10: 0: 0 4% free RaA	0.10			1.1
	10: 9: 6: 4 9% free RaA	0.58			1.6

distribution among the daughters. To illustrate the variation with degree of disequilibrium, the calculations of Harley and Pasternack in Table III-1 yield 0.24 rad/WLM for the equilibrium mixture of Rn and its daughter products and 1.1 rad/WLM with RaB and RaC absent.

Use of the WLM in conjunction with other measurements was discussed at some length at the Workshop. It was suggested that an additional estimation of the unattached fraction of RaA would be useful, short of a more detailed characterization of the atmosphere and daughter concentrations. There was some consensus that the unattached fractions of RaB and RaC are usually small.

The suggestion was made that the old mine atmospheres be re-created today and appropriate measurements taken to supplement the human WLM exposure data. There was disagreement over whether the existing sampling data under- or overestimate actual exposures. The sampled air was often not that to which workers were exposed. Measurements were made at face positions but frequently without ventilation. Mine atmospheres have changed with changing practices in ventilation and the use of diesel fuel. Therefore, comparisons of old data with new are reliable only to the extent that changes in the composition and kinetics of mine atmospheres can be defined and evaluated.

Although the rad/WLM conversion factors are basic to the dosimetry for radon daughters, risk is the concept of importance, especially in view of the new ICRP recommendations. The determination of risk based on absorbed dose, however, raises questions discussed earlier about local high doses and averages. In addition, questions on the values of alpha-particle RBE's for lung carcinomas remain unsettled.

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5. Animal Experiments

Experimental animal studies with inhaled radon daughters have been conducted principally with rats, dogs, and hamsters. Investigations with simulated uranium mine atmospheres are complicated by the need to sort out the roles of a number of factors--radiation, ore dust, smoking, diesel fumes, radon-daughter attachment levels, etc. Moreover, these factors are not independent. Ventilation, for example, affects aerosol concentration and characteristics as well as the degree of attachment and disequilibrium, which, in turn, affect the composition, deposition sites, and clearance of activity in the lung. Synergistic effects probably also result from the combinations of insults in the exposures. Another consideration is the effect of mouth breathing vs. nose breathing.

Animal studies with inhaled radon daughters are summarized by Stuart in Section IV-2. The presence of carrier dust stands out in influencing attachment, clearance, and tumor site. A dose-rate dependence of tumor formation on carrier-dust characteristics may exist, lower rates being more effective.

The relevance of experiments with radon daughters to problems of plutonium inhalation by man was discussed. For both materials one has animal data that include aerosol characterization, lung dose, pathology, and the observed incidence of various pulmonary carcinomas. The extrapolation of the radon-daughter animal data to the human data provided by studies of the uranium miners has important implications for such an extrapolation of plutonium animal data to man. Plutonium shares some of the same problems of carrier-dust effects on lung deposition and retention as well as common problems involving alpha dosimetry.

6. Regulations, Standards, and Remedial Criteria

As indicated in the Introduction, the Workshop was intended to address both technical and philosophical aspects of radon-daughter dosimetry. Both aspects are important, because the practical application of technical knowledge, including its limitations and uncertainties, to control hazards to man involves judgment and a philosophical appreciation of the objectives to be achieved. Radon daughters are probably the major hazard in all radium-bearing wastes. The exposure of populations to elevated levels of radon and radon daughters greatly concerns the public, the Congress, federal state agencies, and various committees that develop and recommend radiation-protection standards. There is an urgent need to develop criteria for guidance in taking some action, or no action, at sites throughout the U. S. where elevated radon concentrations have resulted from man's activities. The criteria will be based on technical knowledge; cost-benefit analyses; and concepts of environmental, social, and legal acceptability.

The history of the interactions between the many federal and state agencies was briefly discussed. A point was made that semantics often confuses an already complicated situation when terms such as "standard" and "regulations" are used. By statute, certain agencies are empowered to issue regulations, which have the force of law, and to provide means and procedures for their enforcement. For example, the Environmental Protection Agency is responsible for establishing occupational WL limits for uranium miners. This agency might also advise states on what levels would be considered as acceptable in other applications, such as for structures built on reclaimed phosphate lands. When federal funds are involved, Congress might specify which agencies have the responsibilities for various phases of remedial action, such as surveys, the development of cleanup criteria, and decontamination.

Although problems about regulations are several steps removed from the technical questions discussed at the Workshop, their mutual interaction is direct. Uncertainties and gaps in scientific knowledge underlie uncertainties about dose conversion factors and risk estimates. This is reflected in the degree to which recommendations and standards are acceptable, usable, and enforceable by regulatory agencies and by those affected. The uncertainties also encourage disagreements when judgments and decisions are made and some action taken. The interplay between the science and the practical world of developing and applying regulations is addressed by Hazle in Section IV-7.

Another aspect of the "science-regulation" interaction involves research. New studies are needed in certain areas and, indeed, are encouraged or supported by agencies with regulatory responsibilities. Some needed research is identified in Section V.

IV. CONTRIBUTED STATEMENTS

1. Summary of Data on Uranium Miners, V. E. Archer

Our prospective study of American uranium miners began in 1950. We added miners to our study group periodically until 1960. We are still following the group and collecting death certificates which are analyzed periodically. Radon-daughter exposure data were collected by various groups over this period. This was incomplete, but has been supplemented by extrapolation and estimates based on average measurements in mining areas. The most recent analyses have been considered in relation to data on underground miners from Canada, Sweden, and Czechoslovakia; considered together, the following relationships have been observed:

a. Lung cancer rates are directly related to cumulative exposure to radon daughters.

b. Lung cancer rates are much higher among smokers than nonsmokers. Amount of smoking has some influence on the lung cancer rate, but it is less marked than the influence of radiation exposure.

c. Lung cancer rates are highest among those who started mining after age 40.

d. The induction-latent period is highly dependent on age at start of mining. The younger a person is when he starts mining, the longer is this period.

e. Lung volume is inversely related to lung cancer rates. In the analyses, height is used as an index of lung volume. The smaller the lungs, the higher the rate.

f. Eye color, as an index of hereditary factors, is also related to lung cancer rates. Miners with blue eyes have least cancer; those with brown eyes have most.

g. The exposure-response curve is not straight. It has a steeper slope at the low-exposure end than at the high-exposure end.

h. Calculations of cancer rate/WLM/10,000 miners indicate that as the dose rate approaches the background level, either alpha radiation is 5 to 10 times more effective in producing lung cancer than at higher levels, or a higher fraction of radon daughters passes through the upper airways at low concentrations.

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i. An altitude effect is likely. The mechanism of the lung volume relationship is probably that small men must ventilate their lungs more when doing the same amount of physical labor than do larger men. Any factor, such as altitude or oxygen deficiency, that causes lung ventilation to increase will almost certainly result in increased lung doses and cancer rates.

j. Most lung cancer in uranium miners arises in the second to sixth bronchial branches. A few (about 10%) arise in more distal areas.

k. The predominant histological type of cancer among the miners is small cell undifferentiated. However, among older men (past 65) there is a tendency for epidermoid types to predominate.

2. Experimental Studies of Inhaled Radon Daughters, B. O. Stuart

Several studies of the biological effects of inhaled radon and radon daughters in experimental animals have been undertaken to determine dose-response relationships under controlled exposure conditions and to define the correlation between exposure and absorbed dose in respiratory tissue.

At the University of Rochester, rodents and dogs have received inhalation of 1800 WL radon daughters without carrier dust, receiving total exposures ranging from 200 to 10,000 WLM. Destructive, hyperplastic, and metaplastic lesions appeared in the lungs, but no tumors. After irradiation ceased, lesions in the bronchial tree were often quickly repaired.

At Limoges, France (Chameaud et al. 1974), rats developed lung cancer between 12 and 24 months after having received 3 to 4 months of daily 4 to 5 hour exposures to radon daughters, with and without uranium ore dust, totaling 500 to 14,000 WLM. Observed pulmonary cancers have included epidermoid (squamous) carcinoma, bronchiolar-adenocarcinoma, and bronchiolo-alveolar adenocarcinoma.

At Battelle Pacific Northwest Laboratories, daily lifespan inhalation exposures of rodents and dogs to 500 WL radon daughters with uranium ore dust at 15 mg/m^3 have produced pulmonary fibrosis and emphysema. Recent findings include squamous carcinoma and bronchioloalveolar carcinoma in 25% of dogs receiving 5 to 6 years of daily 4-hour exposures to radon daughters with uranium ore dust, totaling radon daughter exposure doses of 10,000 to 13,000 WLM. In addition, SPF rats that daily inhaled radon daughters with uranium ore dust totaling 9,000 ± 3,000 WLM have shown metastasizing squamous cell carcinoma, adenocarcinoma, and undifferentiated sarcoma in 60% of those receiving 5-1/2 months of daily exposures. The carcinogenic effects in both large and small experimental animals observed in the Battelle Pacific Northwest Laboratories studies demonstrate a direct cause and effect relationship between lung cancer and inhaled uranium mine aerosols, providing a basis for determining the absorbed radiological dose involved in radon-daughter-induced lung cancer.

Studies are under way to determine the role of concomitant exposures to uranium ore dust as a possible modifying factor in uranium mining carcinogenesis. Recent studies at Limoges, France, of pulmonary carcinogenesis in rats show similar incidences of epidermoid carcinoma after 9,500 WLM of radon daughter exposure without ore dust. However, studies at Battelle Pacific Northwest Laboratories of radon daughters without ore dust show greater nasopharyngeal hyperplasia, but only one or two cases of squamous carcinoma compared to the 60% incidence for radon daughters inhaled in conjunction with ore dust. These findings, together with the fact that studies at the University of Rochester showed minimal changes in doses of 200 to 10,000 WLM radon daughters alone (received in 50 days), compared to the massive pulmonary fibrosis, emphysema, and respiratory tract squamous carcinoma at exposures of 10,000 to 13,000 WLM radon daughters with ore dust observed after several years in the Battelle Pacific Northwest Laboratories studies indicate that:

a. The presence of the ore dust provides physical or physiological interaction that may markedly potentiate carcinogenesis or other pulmonary pathogenesis.

b. There may exist a very significant dose-rate effect, perhaps dependent on recovery mechanisms.

Both possibilities cast doubt upon the adequacy of the cumulative working level month concept as a sufficient index of carcinogenic or other lethal pulmonary disease risk.

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3. Interactions between Radon Daughters and Aerosols,* O. G. Raabe

Abstract

The fundamental considerations relating to radon decay in air and the subsequent adsorption of radon decay products to aerosols are summarized. Since unattached daughters have a different pattern of deposition in the respiratory system from daughters which are attached to aerosol particles, it is important to know the adsorption characteristics and to be able to estimate the fraction of unattached decay products under given circumstances. Also, the aerosol size distribution of the attached fraction will determine its inhalation deposition. Based upon the reported characteristics of radon daughter aerosols in control experiments and in uranium mines, simple relationships are provided for describing the adsorption process and estimating unattached fractions. The principles are described which determine the deposition in the human respiratory tract.

Introduction

Radon gas is formed by the radioactive decay of natural ^{226}Ra as part of the uranium decay series. This gaseous radionuclide may diffuse out of the material matrix in which the radium is found and be transported to the atmosphere. As radon decays by alpha emission in the atmosphere (half-life 3.824 days), the daughter ^{218}Po, called radium A, is formed as a recoiling ion with an initial kinetic energy of 101 keV. It is positively charged when it reaches the end of its recoil path in air of about 100 μm (Hanna 1959). How long it remains charged depends upon the conductivity of the air which depends in part on the concentration of radon and decay products because their emissions cause ionization of air molecules. It is probable that the radium A ion quickly reacts with available oxygen to form polonium oxide (Raabe 1969).

The diffusion coefficient of radium A has been observed to be around 0.05 cm^2/sec (Chamberlain and Dyson 1956). This value is smaller than would be expected for a single uncharged atom, implying either a charged

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atom or small cluster of atoms. Raabe (1968) measured an even lower diffusion coefficient with uncharged radium A at higher relative humidities. The importance of water molecule interactions with radon daughters has not been adequately evaluated, but Raabe (1969) suggested that the formation of a water cluster around the polonium oxide daughter molecule with an effective diameter of 8 Å would explain the observed diffusion rates.

Radium A decays by alpha emission to ^{214}Pb, called radium B, which is initially formed by a recoiling atom. This recoiling atom should undergo reactions which are similar to those of radium A. If the decaying radium A is attached to an aerosol particle, this recoil atom of radium B will penetrate into the particle or jump completely off the particle, depending upon the recoil direction (Raabe 1969). The recoil energy is about 112 keV, so that the recoiling atom will have a range of about 100 μm in air or about 0.025 μm in aluminum. If the radium A is on the surface of the particle and the particle is large (> 0.5 μm), then about half the recoils would be outward; and the recoil fraction, α_a, of radium A which produces unattached radium B would be equal to about 0.5. If the particle is small (< 0.1 μm), it might be expected that the recoiling atom would penetrate through the particle (depending on particle composition and physical density) and would be detached no matter what its recoil direction ($\alpha_a = 1.0$); it might carry along other molecules or particle fragments. If the particle is large but is not a solid, the radium A diffuses into the particle before it decays and the recoil fraction may be much less than 0.5. Mercer (1976) has calculated recoil losses as high as 1.00 or as low as 0.65 for 0.1 μm diameter particles, depending on particle density. Mercer and Stowe (1971) reported an apparent recoil fraction of about 0.81 for radium B recoil from particles with an activity median aerodynamic diameter of 0.2 μm.

Radium B decays by beta emission with a half-life of 26.8 minutes, and its recoil energy is negligibly small. Its decay product is ^{214}Bi, called radium C, which decays by beta emission to ^{214}Po, called radium C'. The half-life of radium C is 19.7 minutes. Since radium C' is an alpha emitter with a half-life of only 1.6×10^{-4} seconds, the radium C beta emission and the radium C' alpha emission are almost simultaneous.

When the primary radon decay products (Fig. IV-1) radium A (^{218}Po), radium B (^{214}Pb), radium C (^{214}Bi), and radium C' (^{214}Po) are free in air as isolated ions, simple molecules, or associated with very small clusters of molecules, they are called unattached (free or uncombined) radon daughters. In contrast, when radon decay products come in contact with the surfaces of the available aerosol particles and condensation nuclei (whose sizes are at least an order of magnitude larger than the unattached daughters), they are said to be attached (or combined).

Radon Daughter Aerosols

Raabe (1969) provides a simple direct method for describing the degree of attachment of radon decay products to aerosols. There are six important reasonable assumptions made: (1) The degree of electrostatic charge of unattached daughters is low since the radon concentrations of biological importance are relatively high, (2) losses to fixed surr. es (such as walls) are not included based upon the assumption that spaces are much larger than a 2-m-diameter cylinder (Raabe et al. 1971), (3) the mass of an unattached radon daughter is negligibly small compared to the mass of an aerosol particle, (4) the diameter of an unattached radon daughter is negligibly small compared to the diameter of a particle, (5) the concentration of unattached daughters with respect to the aerosol concentration is such that there is very little agglomeration of unattached daughters with themselves, and (6) the radon gas concentration does not change during the time periods of interest.

When radon gas enters the atmosphere or is cleared of daughters by filtration, agitation, or other means, the build-up of daughters is limited by the relatively slow decay rate of radon which is long compared to the time period of interest. With N_a and N_r the number concentration in the atmosphere of radium A atoms and radon atoms, respectively; λ_a and λ_r the decay constants of radium A and radon, respectively; and t the time after appearance of daughter-free radon, the appearan. of radium A is given by:

$$N_a = \frac{\lambda_r N_r}{\lambda_a} (1 - e^{-\lambda_a t}). \quad (1)$$

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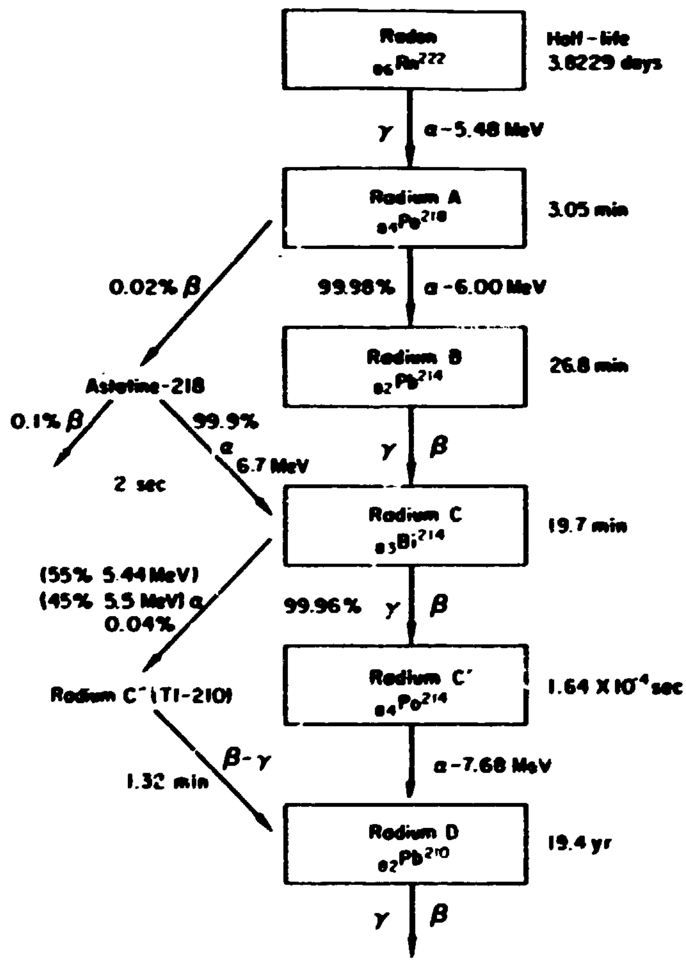

Fig. IV-1. Radon decay chart.

With N_b the number concentration of radium B atoms and λ_b the decay constant for radium B, the appearance of radium B is given by:

$$N_b = \frac{\lambda_a \lambda_r N_r}{\lambda_b(\lambda_a - \lambda_b)} \left[(1 - e^{-\lambda_b t}) - \frac{\lambda_b}{\lambda_a}(1 - e^{-\lambda_a t})\right] . \qquad (2)$$

With N_c the number concentration of radium C atoms and λ_c the decay constant for radium C, the appearance of radium C is given by:

$$N_c = \frac{\lambda_r N_r}{(\lambda_a - \lambda_b)} \left[\frac{\lambda_a}{\lambda_c}(1 - e^{-\lambda_c t}) - \frac{\lambda_b}{\lambda_c} \times (1 - e^{-\lambda_c t}) - \frac{\lambda_a}{(\lambda_c - \lambda_b)}(e^{-\lambda_b t} - e^{-\lambda_c t}) + \frac{\lambda_b}{(\lambda_a - \lambda_c)}(e^{-\lambda_c t} - e^{-\lambda_a t})\right]. \qquad (3)$$

It is useful to define an attachment velocity (rate constant) analogous to the decay constants to describe the rate of adsorption of unattached radon daughter to aerosol particles. The attachment velocity, λ_s, is given by:

$$\lambda_s = \int_0^\infty n\beta(D)P(D)dD \qquad (4)$$

with n the number concentration of particles, $\beta(D)$ the attachment coefficient for particles of size D, and P(D) the aerosol size distribution (probability density). The total adsorption rate of unattached daughters whose number concentration is N_f is given by:

$$\phi_T = \lambda_s N_f . \qquad (5)$$

Employing the attachment relationship given by Raabe (1969) with N_{af}, the number concentration of unattached radium A atoms:

$$\beta(D) = \pi D^2 \bar{v}/4 \qquad (6)$$

for spherical particles of diameter, D, and

$$\lambda_s = S\bar{v}/4 \qquad (7)$$

with S the surface area concentration of the particle, λ_s the attachment velocity, and $\bar{v}$ the average velocity of the unattached daughters ($\bar{v}$ = 1.38 x 10^4 cm sec^{-1}).

The surface area concentration can be described by:

$$S = n\pi\bar{D}_s^2 \tag{8}$$

with n the number concentration of aerosol particles per unit volume of air and $\bar{D}_s$ the diameter of average surface of the aerosol particles for a given distribution of particles.

The concentrations of unattached radium A, unattached radium B, and unattached radium C(N_{af}, N_{bf}, N_{cf}, respectively) can be derived from the differential equations:

$$\frac{dN_{af}}{dt} = \lambda_r N_r - (\lambda_a + \lambda_s)N_{af} \tag{9}$$

$$\frac{dN_{bf}}{dt} = \lambda_a N_{af} + \alpha_a \lambda_a (N_a - N_{af}) - (\lambda_s + \lambda_b)N_{bf} \tag{10}$$

$$\frac{dN_{cf}}{dt} = \lambda_b N_{bf} + \alpha_b \lambda_b (N_b - N_{bf}) - (\lambda_s + \lambda_c)N_{cf} \tag{11}$$

with α_a the recoil fraction of attached radium A which becomes unattached radium B upon decay and α_b the recoil fraction of attached radium B which becomes unattached radium C upon decay. (Indications are that α_b = 0.)

These equations are solved by Raabe (1969).

With these solutions the unattached fractions f_a, f_b, and f_c of radium A, radium B, and radium C, respectively, can be calculated:

$$f_a = \frac{N_{af}}{N_a} = \frac{\lambda_a(1 - e^{-(\lambda_a+\lambda_s)t})}{(\lambda_a + \lambda_s)(1 - e^{-\lambda_a t})} \tag{12}$$

$$f_b = \frac{N_{bf}}{N_b} \tag{13}$$

$$f_c = \frac{N_{cf}}{N_c} \tag{14}$$

Note that the unattached fractions are independent of the radon concentration (the factors $\lambda_r N_r$ cancel in the ratios). For equilibrium times that are large (t > 100 min) the unattached fractions approach:

$$f_a = \frac{\lambda_a}{\lambda_a + \lambda_s} \quad (t \rightarrow \infty) \tag{15}$$

$$f_b = \frac{\lambda_b}{(\lambda_b \quad \lambda_s)} [f_a(1 - \alpha_a) + \alpha_a] \quad (t \rightarrow \infty) \tag{16}$$

$$f_c = \frac{\lambda_c}{(\lambda_c + \lambda_s)} [f_b(1 - \alpha_b) + \alpha_b] \quad (t \rightarrow \infty) \tag{17}$$

Examples

In order to evaluate the distribution of radon daughters adsorbed to aerosols and the unattached fraction for each daughter, it is necessary to know the size distribution of the aerosol as well as the number concentration of aerosol particles. Raabe (1969) used a simple model of the size distribution of atmospheric aerosols to obtain an estimate of the surface area concentration as a function of particle number concentration, n:

$$S = 0.02 \times 10^{-8} \times n \ (cm^{-1}) \tag{18}$$

based upon a $\overline{D}_s$ of 0.08 μm. Different values of S can be calculated from Eq. (18) as required. On the basis of this estimate, the unattached fractions and other characteristic values are shown in Table IV-1.

The most extensive measurements of the activity distribution with respect to aerosol particle size and particle concentration have been reported for uranium mine atmospheres by George et al. (1975). They found activity median aerodynamic diameters between 0.08 μm and 0.34 μm. When all the data were plotted as uncombined fraction versus particle concentration, the results showed considerable spread which can be attributed to the differences in size distribution (Fig. IV-2). The models of Mohnen (1967) and Raabe (1968) have been superimposed upon the data. These models depend upon the aerosol size distribution. The differences between data and models are in part caused by the fact that the data involve a spectrum of size distributions and the models each use but one. However, the model

Table IV-1. Radon Daughter Parameters for Hypothetical Aerosol for Various Aerosol Concentrations n per cc*

n	λ_s	f_a (t = 10 min)	f_b (t = 10 min)	f_c (t = 10 min)	f_a (t = 40 min)	f_b (t = 40 min)	f_c (t = 40 min)	f_a ($t \to \infty$)	f_b ($t \to \infty$)	f_c ($t \to \infty$)
10^3	0.04	0.89	0.83	0.68	0.85	0.54	0.40	0.85	0.36	0.17
5×10^3	0.21	0.57	0.42	0.25	0.53	0.14	0.04	0.52	0.08	0.01
10^4	0.41	0.40	0.24	0.12	0.36	0.07	0.01	0.36	0.04	0.00
5×10^4	2.07	0.11	0.05	0.01	0.10	0.01	0.00	0.10	0.01	0.00
10^5	4.14	0.06	0.02	0.00	0.05	0.00	0.00	0.05	0.00	0.00
5×10^5	20.70	0.01	0.00	0.00	0.01	0.00	0.00	0.01	0.00	0.00
Equilibrium ratios:		1:1.0:0.14:0.02			1:1.0:0.6:0.3			1:1:1:1		

*For these computations, recoil fractions were taken as $\alpha_a = 0.5$ and $\alpha_b = 0$.

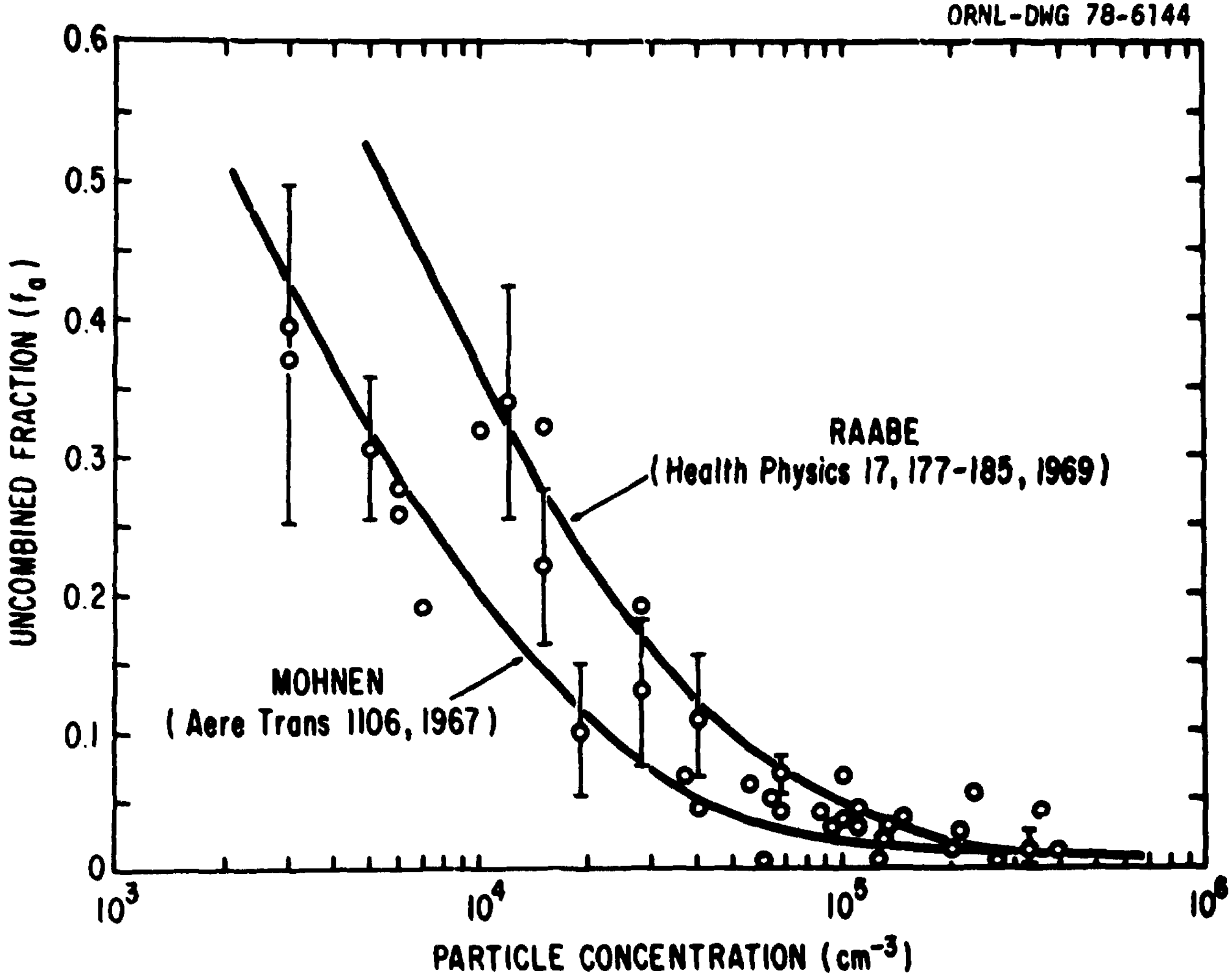

Fig. IV-2. Variation of uncombined fraction of RaA with aerosol particle concentration as determined by George et al. (1975) with the predictions from the models of Mohnen (1967) and Raabe (1969).

of Raabe (1969) is based upon $\overline{D}_s$ = 0.08 μm which is close to the smallest aerosols measured by George et al. (1975). It is appropriate and confirmatory that it described the highest unattached fractions among these data.

Inhalation Deposition

Many models have been proposed for the determination of the dose to the bronchial epithelium of the human lung after inhalation of radon decay products. These have been summarized and discussed by Walsh (1970). These models depend upon choice of particle size distribution, prediction of deposition efficiencies in the human airways during inhalation, and calculation of the dose to certain sensitive basal cells below the epithelial surface. Since most of the radon decay products are usually associated with aerosol sizes under 0.5 μm (including unattached daughters), the primary mechanism of inhalation deposition is Brownian diffusion. For these small particles all of the models use the Gormley and Kennedy (1949) equations which describe diffusional deposition under ideal laminar flow conditions in circular cylinders. The shortcomings of this approach need to be more fully evaluated.

Summary

A simple model has been described which allows the calculation of the radon-daughter activity-size distribution of natural aerosols when the aerosol size distribution and particle number concentration are known. This model agrees well with the best available data on the size distribution of radon daughters in uranium mine atmospheres with respect to estimation of unattached fractions for radium A. Since models of deposition depend upon the activity size distribution of the radon daughters and particularly upon the unattached fraction, these need to be carefully considered in the deposition calculations used to estimate tissue doses.

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4. Is the WLM a Good Index for Risk?, N. H. Harley

During the Workshop there was considerable discussion, if not disagreement, over the quality of the working level month (WLM) as an indicator of risk for radon daughters. It was recognized that the fundamental risk of radon daughter exposure is the alpha dose delivered to bronchial epithelium. This dose is delivered when the daughters establish themselves in the mucous layer through inhalation, deposition, clearance, build-up and decay. Although the physical dosimetry requires detailed characterization of these parameters, nevertheless it is the dose which should be equated with risk. The working level month characterizes only the total alpha energy from radon daughters present in the inhaled air and is therefore deficient in this assessment of dose.

The major benefit of the WLM lies in the extreme simplicity of the measurement, but simplicity is not a substitute for rigor. The WLM can be evaluated quickly and accurately even in the most difficult field conditions. In contrast to this, the atmospheric measurements necessary to perform bronchial dose estimates include the unattached or free ion fraction of RaA, ambient particle size and the concentrations of RaA, RaB and RaC. Unattached RaA is quite important in bronchial dose calculations since these ions deposit with 100% efficiency in the upper respiratory tract, and even a few percent of the RaA which is unattached can contribute a significant fraction of the dose. Otto Raabe pointed out that the free ion component may be calculated easily and reasonably well from a measurement of condensation nuclei if the particle size is known and remains fairly constant.

ICRP recognizes the importance of unattached RaA and includes a correction for its effect in the permissible level for ^{222}Rn. The WLM includes no correction for this effect.

For past mining experience, only WLM is available as an exposure index, so any dose estimates for these atmospheres suffer from lack of any direct measurements. Evaluation of risk in this case in terms of WLM is purely a matter of necessity. Any attempt to evaluate the rad/WLM in these exposures is at the mercy of the various assumptions that must be made.

A skeptical approach to the evaluation of the bronchial dose is usually taken. Even if all of the required atmospheric characterization is performed, there still exists perhaps as much as an order of magnitude uncertainty concerning the lung modeling. The most critical parameter is lung clearance which, if impaired or nonexistent, could easily lead to this large a factor in local bronchial dose rates. The only answer to this is that if a dose-response for radon daughters rather than an exposure-response is ever to be derived, the roots will always lie in this type of calculation. It is fairly certain that a measured dose to cells in bronchial epithelium is impossible. Therefore, we ought at least to calculate the average bronchial dose and then estimate a possible minimum and maximum.

If you must measure WLM, at least obtain either a measurement or an estimate of unattached RaA. This would allow a beginning to the problem of realistic dose evaluation.

5. Additional Comments on Radon-Daughter Dosimetry Models, V. E. Archer

I seem to have more reservations about the accuracy and applicability of dose calculations for radon daughters to the bronchi than do the physicists. The reservations are based on several considerations:

a. Free ions in most of the calculations deliver a disproportionate amount of the dose to the bronchial mucosa. However, a much higher fraction of free ions is removed in the nasopharynx, trachea, and large bronchi than of attached daughters. Because of thickness of mucosa and depth of basal layers, the deposition in these areas is harmless. This ineffective fraction may be 40-99% of the free ions and varies much more in mouth vs. nose breathing than do other fractions of radon-daughter-carrying particles. I think the analogy between free ions and SO_2 absorption in the nasopharynx and trachea is probably pretty good. In fact, one would suspect that a higher fraction of free ions than SO_2 would be removed. Several experiments in man and animals with SO_2 have shown that from 90 to over 99% of SO_2 is removed in the nose. The actual fraction removed varies a great deal with humidity, with inert particle size and abundance (the more smaller ones, less than 1 μm, the greater the SO_2 penetration), with respiratory rate and with absolute concentration of SO_2 (a higher percent is removed at higher concentrations up to certain levels). I am not at all sure that George's experiments with retention in room-type air are applicable to mines.

b. Humidity varies greatly in mines and with ventilation. The higher the humidity and the "older" the radon daughters, the larger are the "free ions" and radon-daughter-bearing particles. This tendency for charged particles to enlarge is opposed by the "settling out" of larger particles in "old" air--leaving only the smaller ones suspended. Actual size distribution of radon-daughter-bearing particles is therefore not a constant as visualized by the models.

c. All radon daughters bear charges when formed. These charges cannot be fully neutralized by attachment to dust particles. The older the air, the more likely the charges are to be neutralized. There is also evidence that "free ions" quickly become oxidized and coated with water molecules. Therefore, there should be no sharp demarcation between "free" and "attached" radon daughters. Instead, there is a continuous spectrum of particle sizes bearing various electrical charges. The larger the particle, the less the influence of the charge on particle movement. The distribution within this spectrum must vary a great deal, depending on age of the air, humidity, and turbulence. Although the movement of particles as large as 1 μm in diameter may not be influenced much by the charge, movement of many of the smaller-sized particles probably is. It is therefore a great oversimplification to regard this spectrum as having only two components: "free" and "attached" daughters. In fact, this terminology has arisen as a result of instrument limitations--separation of only the smallest fraction. However, it is apparent that different techniques for measurement of "free ions" measure different fractions of the spectrum. In addition there is a small charge on bronchial walls to further confuse things. The presence of charges on particles influences particles in several ways: (1) by interaction between particles--repulsion or attraction [Repulsion would tend to keep particles small; attraction would make them larger by agglomeration] and (2) by being attracted to or repelled by the mild charge of the respiratory mucosa.

d. The thickness of bronchial epithelium is not constant throughout the tracheobronchial tree. It is thickest in the trachea and gradually thins out to a single layer of cuboidal cells in the bronchioles. Calculations of dose only to the basal layer of epithelium in the mid-size bronchi may therefore be misleading when those calculations are used for averaging throughout the bronchial tract.

e. The effects of turbulent air flow in the tracheobronchial tree have, I think, been greatly underestimated by the physicists, possibly because they don't know how to deal with it. During respiration the bronchi are constantly changing in size. Their diameter is maximum at inspiration and minimal at expiration. In addition, the many curves and divisions in the bronchial tree, along with the periodic reversal of flow direction, all cause turbulence. Turbulent air flow in the nose and bronchi must be the rule rather than the exception. Turbulence tends to negate the applicability of diffusion constants.

Summary

The factors mentioned above which have not been adequately considered in the lung models lead to the following conclusions: (a) "free ions" due to their large removal in the upper airways and their rapid enlargement due to oxidation and water absorption may not be as important as considered in many models, (b) electrical charges on most radon daughters and turbulence of air flow probably result in greater deposition of "attached" radon daughters in the 2 to 6 bronchial generations than is calculated by most models, and (c) calculations of rad dose are considerably more uncertain than many physicists contend.

I also have fewer reservations about variation in the rad value of the WLM than do many of the physicists. Although this conversion is capable of large theoretical variation due to variations in "free ions" and radon daughter ratios, I don't think its variation in actual mines is very great. My reasons for this are: (a) the factors noted above which minimize the importance of "free ions" and (b) the measurements of Raghavayya and Jones (1974) which were made in mines, representing a wide variety of ventilating conditions. They explored the relationship between free ions, radon daughter ratios, and dust with ventilation. Their findings indicate that increasing ventilation in mines over the years probably has not greatly changed the fraction of free ions or radon daughter ratios. The subsequent letter to the Editor of Health Physics pointing out an error in their calculations does not alter the above conclusion.

Reference

Raghavayya, M., and Jones, J. H., 1974, "A Wire Screen-Filter Paper Combination for the Measurement of Fractions of Unattached Radon Daughters in Uranium Mines," Health Phys. 26, 417.

6. A General Comment on Risk Evaluation, P. G. Groer

While I share the concerns of many participants about the WLM-to-dose conversion factor, I am bothered more by two problems in determining the ordinate of the dose-response (or exposure-response) curve. First, time is usually not considered despite its tremendous importance for the shape of the dose-response curve. To illustrate this point, consider the examples shown in Figs. IV-3 and IV-4. At the larger dose, D_2, the individuals at risk may not live long enough to show a response higher than at dose D_1 (Fig. IV-3) or the response at D_2 would increase further (Fig. IV-4). As far as I know, usually, only the fraction of miners showing lung cancer is plotted versus dose regardless of the total time at risk in the different dose groups. The second problem with the ordinate stems from the fact that miners die not only from lung cancer but also from many other causes. The information that some miners lived for a certain number of years and then died from other causes is, again, not used in the "crude" dose-response curve which is usually plotted. As in some animal studies, competing risk theory (David and Moeschberger, 1978) should also be used for human epidemiological studies. This would lead to a unified methodology and a meaningful comparison of lung cancer incidence in uranium miners in different countries.

Reference

David, H. A., and Moeschberger, M. L., 1978, The Theory of Competing Risks, to be published by Griffin and Co.

ORNL-DWG 78-6146

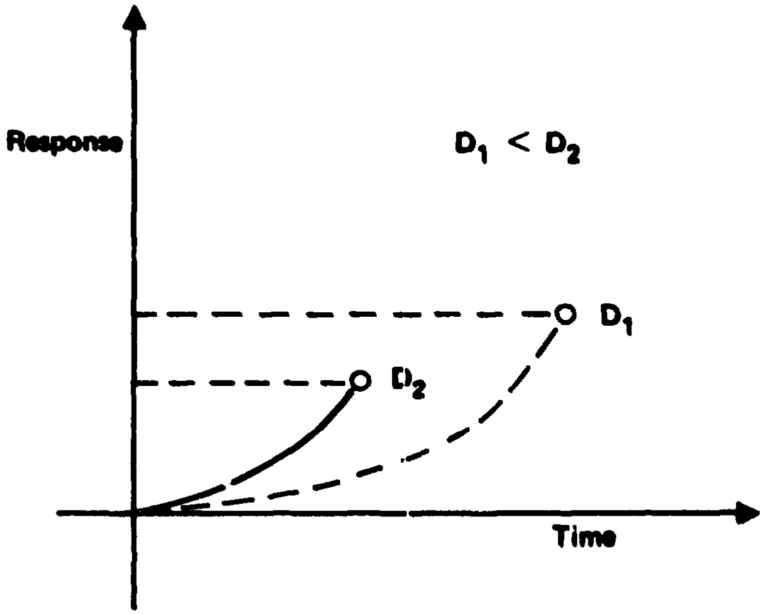

Fig. IV-3. Dose-response curves and time. The response at the higher dose D_2 stayed below the response at the lower dose D_1 because of enhanced life shortening at the higher dose level.

ORNL-DWG 78-6145

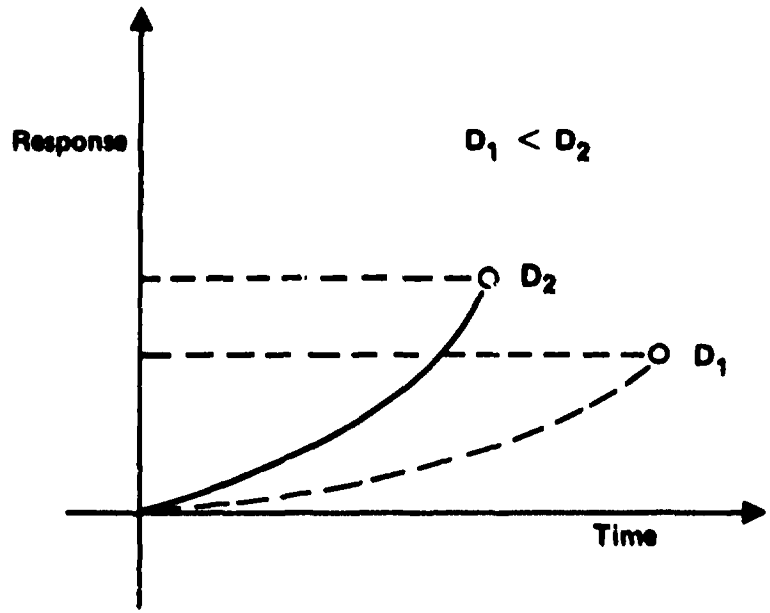

Fig. IV-4. Dose-response curves and time. The response at the higher dose D_2 is greater than the response at the lower dose D_1 and would have increased further if enhanced life shortening would not have interfered.

7. Practical Application: Radon-Plus-Daughters Dosimetry and Regulations, A. J. Hazle

Historically, radon has been an occupational problem in some types of mines. As early as the 1500's, miners in the Erz Mountains in Europe were known to have a high incidence of total lung disease. The mines in which they worked produced copper, iron, silver, and other metals. In the late 1800's, the mines produced pitchblende for dyes and later for uranium and radium. By the late 1920's, it was established that the cause of death was really lung cancer and not lymphosarcoma or tuberculosis as earlier suspected. About 50% of the deaths observed were due to lung cancer.

Measurements made in the mines during the late 1920's identified high levels of radon gas. It was not until 1965 that studies made in the 1950's and 1960's on the Colorado Plateau miners allowed the Public Health Service to show a dose-response relationship between cumulative radon daughter exposure and the incidence of lung cancer. The National Academy of Sciences BEIR Report of November 1972 used 5 rem per CWLM (cumulative WLM) as a dose conversion term. Currently, the Environmental Protection Agency is using 16 rem/CWLM.

Studies of radon daughter inhalation exposure in animals were uniformly unsuccessful in demonstrating the relationship between radiation exposure and lung cancer until French researchers identified a quantitative dose-response relationship in 1975. This research provides sufficient evidence that radon daughter inhalation exposure can cause lung cancer, although other materials in the mine air may be contributory.

One of the problems in establishing a dose-response relationship for nonoccupational exposure to radon daughters is that the unit of concentration is the working level (WL). Many factors influence the radiation resulting from a given numerical value of WL. Some of the most prominent are the radon gas concentration, equilibrium of radon daughters, and the relative humidity of the air. In a mine these factors are usually fairly constant, but in the general environment they vary widely. However, the average annual value of working levels (CWLM) in the environment is a good estimator for a dose-response evaluation from a practical health physics standpoint.

A second problem in evaluating the dose-response relationship is that miners and a general population have a different physiological make-up. The major factors are age, sex, and the condition of the respiratory system. The last factor includes the effects of smoking and of working in a mine atmosphere. (Miners may have a thicker mucous lining of the respiratory tract because of the contaminants in the mine air.)

The radon-daughter dose is delivered to the bronchial epithelium of the lung, which is much more sensitive to radiation compared to the pulmonary region. Because bronchial cancer (the uranium miner lung cancer "type") is believed to have many possible causes, the bronchial cancer risk from radon-daughter exposure is best expressed in terms of the percentage increase in the expected cancer risk. The potential risk is assumed to vary in direct proportion to the radon-daughter concentration in units of CWLM. EPA has recently recommended the use of 55 CWLM as the exposure assumed to double the risk of bronchial cancer for members of the general population. (The figure 110 CWLM is used for estimating uranium miner lung cancer risk.) Therefore, an individual exposed to 0.01 WL continuously accrues 0.5 CWLM/year, which is equivalent to a 1% increase in potential risk for each year of exposure, or a 70% increase in potential risk over a lifetime of 70 years. The above rate is consistent with national vital statistics and natural background radon-daughter levels.

Because of the problems associated with the estimates of probable effects of radon-daughter exposure, only basic statements can be made. However, it is assumed that:

a. Radon-daughter exposure can induce lung cancer.
b. Depending on the risk of lung cancer in any age group, radon-daughter inhalation above background will increase the risk by a certain percent.
c. The risk of lung cancer, based upon the uranium miner experience and adjusted for general population application, is about 1% per CWLM of exposure; 100 CWLM will double the natural risk.

Thus, it can be seen that there is justifiable concern for exposure to elevated radon-daughter exposures identified for certain populations.

Consideration of these scientific data in the enactment of legislation and in subsequent actions is seen in the Grand Junction Remedial Action Program. In October 1971, the Subcommittee on Raw Materials of the Joint Committee on Atomic Energy of the U. S. Congress held hearings in Washington, D. C., regarding the use of uranium mill tailings for construction purposes in Grand Junction. As a result of these hearings, on June 16, 1972, Public Law 92-314 was enacted authorizing appropriations for assistance to the State of Colorado in initiating remedial action on the basis of the U. S. Surgeon General's Guidelines.

Subsequent to Public Law 92-314, the Atomic Energy Commission (AEC) regulations were amended in 1972, and in accord with 92-314, to establish the Grand Junction Remedial Action Criteria and was known as 10 CFR 12. With the demise of AEC, the legal regulatory reference is now 10 CFR 712 and the contract between the State of Colorado and the Federal Government (Energy Research and Development Administration, ERDA; now Department of Energy) for the conduct of the remedial program.

In addition to the standard portions of such regulations, 10 CFR 12 contains general radiation exposure level criteria for remedial action as found in the Surgeon General Guidelines (12.6); criteria for determination of possible need for remedial action where criteria have not been met (12.8); factors involved with priority of remedial action (12.9); and the selection of appropriate remedial action (12.10).

While 12.6 gives the basic philosophy and 12.7 gives the "absolute" numerical values, 12.8 provides for mitigating circumstances which must be considered if the program is to be truly effective. Needless to say, the mitigating circumstances are difficult to describe as to the effect involved. However, several have been demonstrated to be quite significant in their impact, such as a change in the structure's use.

The Mine Enforcement and Safety Administration and the Occupational Safety and Health Administration also have regulations which govern occupational exposures as does the Nuclear Regulatory Commission (NRC) and the Agreement States in their respective jurisdictions. Primarily, these are based on the recommendations of the NCRP and not on recent health risk evaluations.

Let us now turn to estimates of the Grand Junction remedial program effectiveness. The program had effectively completed remedial action on 200 occupiable structures as of July 1976, which included 187 residences, 11 schools, and 2 commercial structures. The number of persons affected by the remedial action was 5,584, with 731 in residences, 4,841 in schools, and 12 in commercial buildings.

By individual location evaluation, the gamma and radon daughter dose reductions have been estimated. The overall average annual gamma dose reduction per occupant has been 0.051 rem, with 0.172 as the average for residences, 0.033 for schools, and 0.040 for commercial buildings. The overall annual average radon daughter dose reduction per occupant has been 9.3 rem, with 64.4 rem for residences, 1 rem for schools, and 14.3 rem for commercial structures. The construction cost of remedial action on these 200 locations was $3,316,097, of which $2,100,168 was spent on residences, $1,187,366 on schools, and $28,563 on the commercial structures.

When these cost and dose reduction figures are combined, the overall average dose reduction cost is found to be $63/rem, with $44/rem for residences, $243/rem for the schools, and $166/rem for the commercial buildings.

The above data are preliminary, as a total of about 600 locations are estimated to be in need of remedial action.

Although the situation does not truly lend itself to the man-rem concept due to the nonuniformity of exposure, the 9.3 rem/year exposure of the 5,584 occupants based on a 16 rem/CWLM conversion would be equivalent to a 1% increase in potential risk of lung cancer per year to the population so exposed on a relative risk basis. The absolute risk concept as described in the BEIR Report would provide even higher estimates of risk. Due to the nature of nonuniformity of exposure, the risk would be necessarily lower than the 1%/year estimate.

In October 1974, a summary report was issued on the Phase I study of Inactive Uranium Mill Sites and Tailings Piles. This study was requested of EPA and ERDA by Congress. Phase I involved site visits to determine site condition, need for corrective action, ownership, proximity to populated areas, and aspects for increased population near the site. The summary of the Phase I report states that the inactive uranium mill tailings pile stabilization work done as required by Colorado "represents a holding action, sufficient for the present, but not a satisfactory answer for long-term storage."

A Phase II report has been issued by the ERDA contractor on the Salt Lake City pile. Data acquisition on the Colorado tailings piles is in its final stages, and published reports on the individual sites should be available in the next several months. Phase II of the study includes evaluation of the problems and examination of alternative solutions, the preparation of cost estimates, and detailed plans and specifications for alternative remedial action measures.

Over the past several years, EPA has conducted its own investigations on the hazards associated with tailings piles. Reports have been issued regarding the radon emanating from the surface of the piles and gamma surveys in the area of uranium tailing piles. Most recently, a report was published on the potential radiological impact to individuals living near tailings piles. Concern is expressed for the radon daughter concentrations which may be observed in a structure close to the pile due to ambient (outside air) levels provided by the presence of the pile itself. The Phase II EPA/ERDA study on the situation will provide more information and cite specific data in this regard. A 1974 Outdoor Radon Study was conducted by EPA and Colorado, and a published report is anticipated in the very near future.

As is evident, Colorado and other associated agencies have spent a great deal of time and money in investigating problems associated with mill tailings and remedial actions. Considerably more effort will be required before the entire situation is finally resolved. Also, NRC is in the process of developing a Generic Environmental Impact Statement on uranium milling and will address some of these topics to preclude future problems.

A word about regulations. Technical regulations are complex and a bore. Everyone can find fault and is dissatisfied. Generalized regulatory statements are unmanageable and open the agency to additional criticism due to ambiguity. Somewhere in between is an optimum probably generally acceptable to the regulator and regulatee. While no regulatory statement will be absolutely correct due to changes in regulatory philosophy and changing technical knowledge, the statement must be reasonable, conservative, and so stated that any error made in the health physics aspects would not be grossly in error for the generalized circumstances covered in regulation. The technical verbiage must fit the practical regulatory effort to accomplish the desired protection of the worker, the general population, and the environment.

References

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V. WORKSHOP OBSERVATIONS AND RECOMMENDATIONS

J. E. Turner and C. F. Holoway

As stated in Section I-1, the objectives of the Workshop were to provide an opportunity for the exchange of information and to develop this report, assessing technical problems in dosimetry for radon and radon daughters. While it cannot be claimed that this document provides a definitive update of the subject, it does focus on certain aspects of radon-daughter dosimetry that reflect current concern. In this section the highlights of the Workshop are summarized.

Table V-1 lists these highlights according to the critical areas discussed and the principal issues involved. Some remarks are given to try to reflect the general tenor of the discussions. Reference is also made in the table to relevant sections of this report.

Aerosols. The production of radon-daughter ions by decay in air and their subsequent fate (further decay, attachment to aerosols or adsorption on other surfaces) are understood in considerable detail. Complicating factors include ventilation, humidity, and chemical composition of the aerosols. In Section IV-3 Raabe has described a model for calculating the unattached fraction and the distribution of attached daughters as a function of aerosol size. While more measurements could be used for additional verification, the treatment of aerosols is probably adequate for most applications, especially when compared with other uncertainties in radon-daughter dosimetry.

Dose Calculations. One of the unresolved problem areas in the dosimetry for radon daughters in the respiratory tract is the choice of the relevant lung-tissue mass to use in specifying dose. This is a long-standing general problem that occurs when the dose distribution from a radionuclide is very nonuniform in tissue. Related examples include plutonium in the lung and bone and radium in the bone. In these and other cases, particularly with alpha emitters, local doses in small volumes of an organ can exceed by many times the average dose to the organ.

Of the various parameters assessed for dose calculations--deposition, clearance, cell at risk--the most poorly known is clearance. The mucous structure as well as its production rate, movement, and different clearance rates in different parts of the lung is poorly known or understood. The

Table V-1. Summary of workshop discussions

Critical area discussed	Issues	Remarks	Sections of this report
Aerosols	Distribution of adsorbed radon-daughter air activity by particle size and, particularly, estimation of unattached fractions.	Understanding is probably adequate.	III-2 IV-3
Dose Calculations	Deposition of particulates in regions of respiratory tract. "High, localized" dose vs. "average" lung dose. Mucous structure; lung clearance.	Average doses probably reliable. Accuracy of calculations limited by biological factors.	II III-3 III-4 IV-5
Rad/WLM Conversion Factors	Variation with type of atmosphere, radioactive disequilibrium factor, distribution of attached and unattached daughters, and lung model used.	For given exposure conditions, conversion factors are in general agreement.	III-4 IV-5 IV-7
Use of WLM or Dose	WLM is tied to human epidemiologic data. Cancer risk results from alpha-particle dose. Rad/WLM depends on distribution of potential alpha energy among daughters.	No general agreement on whether WLM or dose should be considered as primary index of risk.	III-1 III-4 IV-2 IV-4 IV-5
Risk	Knowledge of exposure-response curve; excess lung cancer incidence rate per WLM; dependence on age, sex, and other factors. Effects of smoking and exposure to other concomitant stresses. Existence of dose-rate effects.	Data reviewed. No attempt made to critique risk estimates in current use.	III-1 IV-1 IV-2 IV-6 IV-7
Remedial Criteria	Development and application of suitable criteria for reduction of population exposures to radon daughters.	Concern expressed about validity of applying data for miners to general population. Coupling with socio-economic factors outside scope of workshop.	III-6 IV-7

discrete, rather than continuous, structure of the mucus needs further study. The origin of tumors in regions apparently outside the range of alpha particles needs clarification. While deficiencies in the calculations were recognized, it was generally agreed that computed average doses to the bronchial epithelium are probably reliable.

Rad/WLM Conversion Factors. No attempt was made to suggest a "best" value or range of values to use in relating absorbed dose and working level month, even under specified exposure conditions. A number of calculations and reviews were discussed, as summarized in Section III-4. No strong issue was taken with existing published values, although, as already pointed out, the high value of Haque and Collinson in Table III-1 is the result of assumptions that are not apt to be met in practice.

Use of WLM or Dose. The pros and cons of using the working level month, rather than absorbed dose, as a primary reference level were challenged on both sides by several participants. The variability of WLM-to-dose conversion factors hinges mainly on the unattached fraction, although particle size, unless it has essentially remained unchanged over the history of uranium mining, would also be expected to alter the dose to the tracheobronchial region. It was suggested that some measurement of the unattached fraction along with WLM be made a legal requirement. Some attendees felt that such measurements are unnecessary in many cases. In the majority of mines, for example, the unattached fractions are always $\lesssim 0.1$. There was also considerable sentiment that, while not perfect, the present use of the WLM, tied as it is to the human epidemiological studies, is warranted. However, caution was expressed in predicting risks to future miners, who may be exposed to radon daughters under different conditions than the earlier miners, and to individuals in a population at large exposed to elevated radon-daughter levels.

The possibility of reconstructing old mine atmospheres and carrying out some detailed measurements was considered. Feeling was expressed both ways whether such a program would be useful. A point in question is how representative the measured samples would be of the air actually breathed in the old mines.

Risk. Human experience and data from animal experiments were discussed at some length. The principal relationships that appear from existing data are summarized in Sections IV-1 and IV-2. No attempt was made at the Workshop to assess various risk estimates that have been applied by scientific and government organizations.

Remedial Criteria. Remedial programs, such as that at Grand Junction, have involved a great deal of time, effort, and expense. Decisions must be made about possible actions at a number of inactive and active uranium mill tailings sites. As mentioned just above, concern was expressed about the application of WLM limits, based on studies of miners, to other populations under different exposure conditions. In homes and public buildings, for example, ventilation rates, humidity, and unattached fractions and physical-chemical differences in the carrier aerosols are different from what they are in mines. Other factors, such as the number of persons at risk and cost-benefit analyses, should be taken into account in considering remedial actions.

Indicated Research. Although not included as an item in Table V-1, further research is clearly needed in a number of areas, as evidenced by the numerous questions raised in the Workshop discussions and throughout this report. The most serious deficiencies in radon-daughter dosimetry appear to be in lung-particulate dynamics (turbulence, lung physiology, clearance) and in the specification of dose itself (use of average dose, effects of localized high doses).